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Enclosed please find the final technical report for the Cooperative Agreement #301521 for the period April 1, 2000 to March 31, 2003.

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Title: High Resolution Spectroscopy to Support Atmospheric Measurements

Final Technical Report

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Summary of Research:

A summary of the Research activities performed during the period April 1, 2000 to March 31, 2003 under Cooperative Agreement #301521 by Dr. Malathay Devi Venkataraman (PI) is given in the following pages.

Spectroscopic parameters [such as line position, intensity, broadening and shifting coefficients and their temperature dependences, line mixing coefficients etc.] for various molecular species of atmospheric interest were determined. In order to achieve these results, infrared spectra of several molecular bands were obtained using high-resolution recording instruments such as tunable diode laser spectrometer and Fourier transform spectrometers. Using sophisticated analysis routines [Multispectrum nonlinear least squares technique] these high-resolution infrared spectra were processed to determine the various spectral line parameters that are cited above.

Spectra were taken using the McMath-Pierce Fourier transform spectrometer (FTS) at the National Solar Observatory on Kitt Peak, Arizona as well as the Bruker FTS at the Pacific Northwest National Laboratory (PNNL) at Richland, Washington.

Most of the spectra were acquired not only at room temperature, but also at several different cold temperatures. This procedure was necessary to study the variation of the spectral line parameters as a function of temperature in order to simulate the Earth's and other planetary atmospheric environments. Depending upon the strength [or

weakness] of the various bands recorded and analyzed, the length(s) of the absorption cells in which the gas samples under study were kept varied from a few centimeters up to several meters and the sample temperatures varied from approximately $+30^{\circ}$ C to -63° C. During the Cooperative Agreement [#301521] period, research on several infrared bands of various molecular species and their isotopomers were undertaken. Those studies are briefly descried in the following pages.

Molecules whose spectra were recorded and analyzed consisted of:

Isotopic methane [CH₃D], Ozone [$^{16}O_3$], Ozone isotopomers [$^{16}O^{17}O^{16}O$ & $^{16}O^{16}O^{17}O$], carbon monoxide [CO], Carbon dioxide [CO₂] and hydrogen cyanide [H $^{12}C^{14}N$].

Specific molecular bands analyzed are: The fundamental bands v_3 , v_5 and the v_6 of $^{12}\text{CH}_3\text{D}$; the combination band $v_1 + v_3$ of ozone isotopomers $^{16}\text{O}^{17}\text{O}^{16}\text{O}$ & $^{16}\text{O}^{16}\text{O}^{17}\text{O}$]; the v_3 and the v_1 bands of the main ozone isotopomer $^{16}\text{O}_3$; the first overtone band of the main isotopomer of carbon monoxide [$^{12}\text{C}^{16}\text{O}_2$] and $^{13}\text{C}^{16}\text{O}_2$ isotopomer.

A brief description of the various studies is furnished below.

1. Lorentz self-, and nitrogen-broadening coefficients as well as self-, and nitrogen-pressure induced shift coefficients were measured at room temperature for over 440 transitions in the ${}^{Q}P$, ${}^{Q}Q$ and ${}^{Q}R$ branches of the v_3 (A₁) parallel band of ${}^{12}CH_3D$. The simultaneous multispectrum nonlinear least squares fitting analysis was performed in the 1140-1440 cm⁻¹ region using 14 spectra of room temperature gas recorded with the McMath-Pierce FTS. The measured broadening and pressure-shift coefficients were found to vary with the rotational quantum index numbers J'' and K''. Weak line mixing effects were observed in several high-J lines with K'' = 3 splitting and approximate values for the off-diagonal relaxation matrix element coefficients were determined for a few A_1A_2 split components.

Similar measurements were made for the $v_5(E)$ perpendicular band of $^{12}CH_3D$ located between 1275-1600cm⁻¹ as well as for the $v_6(E)$ perpendicular band of $^{12}CH_3D$ in the 1035-1270cm⁻¹ regions. These studies are important for interpretation of the infrared

spectra of the atmospheres of some of the giant planets and their moons [e.g. Titan]. Knowledge of the spectral line parameters for individual rotation-vibration lines of CH₃D is also important for quantitative spectroscopic analysis of industrial and research gas samples containing methane and its isotopomers.

2. Using very high resolution $[0.002 \text{ cm}^{-1}]$ Fourier transform spectra of a ^{17}O -enriched sample, an extensive analysis of the v_1+v_3 bands of the $^{16}\text{O}^{17}\text{O}^{16}\text{O}$ and $^{16}\text{O}^{16}\text{O}^{17}\text{O}$ isotopomers of ozone has been performed for the first time. Using a Hamiltonian matrix which took various resonance's into account, precise vibrational energies and rotational and coupling constants were deduced, leading to a more accurate band center determination of v_0 (v_1+v_3) =2078.3496 cm⁻¹ for $^{16}\text{O}^{17}\text{O}^{16}\text{O}$ and v_0 (v_1+v_3) =2098.8631 cm⁻¹ for $^{16}\text{O}^{16}\text{O}^{17}\text{O}$. These analyses are important because the spectral features of these isotopomers are used in remote sensing studies of ozone in the troposphere and stratosphere. The complete characterization of ozone absorption spectrum, including isotopic and hot bands, is essential to quantitative analysis of atmospheric absorption and emission spectra.

We have also identified for the first time isotopic ozone lines of $^{16}O^{16}O^{17}O$ and $^{16}O^{17}O^{16}O$ in the 5-µm region. These spectral features were observed in balloon-borne high-resolution (0.003 cm⁻¹) solar absorption spectra in addition to a few lines that were also observed in the ground-based spectra. These lines are important in the analysis of atmospheric absorption spectra.

In addition to the above discussed ozone bands, we have also measured the absolute intensities of 376 lines of the fundamental v_1 and v_3 bands of the main isotopomer, $^{16}O_3$. These two bands fall in the 9 to 11 μ m region. In this experiment, the ozone samples were contained in a glass cell having crossed IR-transmitting and UV-transmitting paths, and the UV absorption at 254 nm was used to monitor ozone concentration in the cell.

3. We made room temperature measurements of self- and hydrogen-broadening coefficients and pressure shift coefficients for the first overtone (2-0) band transitions of ¹²C¹⁶O through the analysis of several high-resolution FTS spectra. This study presents the first high-resolution experimental determination of hydrogen-induced pressure

broadening and pressure-shift coefficients in the 2-0 band of ¹²C¹⁶O. The results from this work should be useful for interpretation of planetary atmospheres data and also provide a useful and reliable dataset for comparison to theoretical modeling of broadening and pressure shift coefficients involving CO transitions.

- 4. We have made the first high-resolution measurements of pressure-broadening and pressure-induced shift coefficients due to nitrogen for a large number of rovibrational transitions in the v_3 band of $^{13}C^{16}O_2$. In addition, nitrogen-broadening coefficients for many transitions in the $v_3+v_2^1-v_2^1$ hot band of $^{13}C^{16}O_2$ as well as the v_3 band of $^{13}C^{16}O^{18}O$ were also determined. These results should be valuable not only for predicting the infrared spectrum of the atmosphere due to CO_2 absorption or emission but also for verifying theoretical calculations of pressure broadening and pressure shift coefficients in the v_3 region of carbon dioxide spectra.
- 5. Extensive high-resolution experimental determination is also made for air- and N_2 -broadening and air- and N_2 pressure-shift coefficients for the two $^{13}C^{16}O_2$ laser bands (located at 913.4 and 1076.6 cm⁻¹, respectively), in addition to new measurements of self-broadening and self-shift coefficients for the two $^{12}C^{16}O_2$ laser bands. By combining the spectra of $^{12}C^{16}O_2$ and $^{13}C^{16}O_2$ in the same least squares fit we were able to obtain a consistent set of line parameters for both molecules. Absolute line intensities were also determined for the two $^{12}C^{16}O_2$ laser bands (v_3 - v_1 and v_3 - $2v_2^0$) centered near 960.9 and 1063.7-cm⁻¹, respectively. Using a multispectrum fitting technique we simultaneously fit 30 spectra in these studies. These results should be included in the future updates of the HITRAN [High resolution TRANsmission] line parameters compilation.
- 6. The infrared spectrum of HCN in the region between 3150 and 3450 cm⁻¹ has been recorded at 0.005 and 0.008 cm⁻¹ resolution using two different Fourier transform spectrometers, the McMath-Pierce Fourier transform spectrometer located at the National Solar Observatory (on Kitt Peak) and the Bruker-120HR Fourier transform spectrometer situated at the Pacific Northwest National Laboratory at Richland, Washington. Room temperature measurements were made of positions, absolute

intensities, self broadening and self shift coefficients for individual lines belonging to the HCN v_1 band centered near 3311 cm⁻¹. These are to our knowledge the first extensive set of self-broadening and self shift measurements in the band. In addition, intensities, self broadening and self shift coefficients for several lines of the $v_1+v_2^1-v_2^1$ hot band and several intensities for lines in the H¹³C¹⁴N and H¹²C¹⁵N isotopomers were also determined. A multispectrum nonlinear least squares fitting algorithm was used to fit the entire spectral region covering the 3200 to 3400 cm⁻¹ region of up to 27 spectra simultaneously.

In addition to intensities, air– and N_2 -broadening, air– and N_2 -pressure–induced shift coefficients and the temperature dependence exponent of the broadening coefficients and the temperature dependent coefficient of pressure-induced shifts have been determined for transitions of the v_1 band of $H^{12}C^{14}N$ from 39 laboratory absorption spectra recorded at 0.005 and 0.008–cm⁻¹ resolutions. Two articles have been submitted to the special issue of J. Quant. Spectrosc. Radiat. Transfer that is expected to be out in the fall of 2003.

The following Journal articles and Oral Presentations resulted from the 3-year Cooperative Agreement period.

Journal Articles:

- 1. Measurements of air-broadening, pressure shifting and off-diagonal relaxation-matrix coefficients in the v_3 band of $^{12}CH_3D$.
 - V. Malathy Devi, D. Chris Benner, M. A. H. Smith, C. P. Rinsland, and L. R. Brown.
 - J. Mol. Structure, 517-518, 455-475 (2000).
- 2. Measurements of air broadened width and air-induced shift coefficients and line mixing in the v_6 band of $^{12}CH_3D$.
 - V. Malathy Devi, D. Chris Benner, M. A. H. Smith, and C. P. Rinsland.
 - J. Quant. Spectrosc. Radiat. Transfer, 68, 1-41 (2001).
- 3. Measurements of air broadened width and air-induced shift coefficients and line mixing in the v_5 band of 12 CH₃D.
 - V. Malathy Devi, D. Chris Benner, M. A. H. Smith, and C. P. Rinsland.
 - J. Quant. Spectrosc. Radiat. Transfer, 68, 135-161 (2001).
- 4. Absolute intensities of O_3 lines in the 9-11 μ m region.
 - M. A. H. Smith, C. P. Rinsland, V. Malathy Devi, and D. C. Benner.
 - J. Geophys. Res. 106, 9909-9921 (2001).
- 5. The v_1+v_3 bands of the $^{16}O^{17}O^{16}O$ and $^{16}O^{16}O^{17}O$ isotopomers of ozone.
 - A. Perrin, J.-M. Flaud, F. Keller, M. A. H. Smith, C. P. Rinsland, V. Malathy Devi,
 - D. Chris Benner, T. M. Stephen and A. Goldman.
 - J. Mol. Spectrosc. 207, 54-59 (2001).
- 6. Multispectrum analysis of self- and N_2 -broadening, shifting and line mixing coefficients in the v_6 band of $^{12}CH_3D$.
 - V. Malathy Devi, D. Chris Benner, Linda R. Brown, Mary Ann H. Smith, Curtis P. Rinsland, Robert L. Sams, Steven W. Sharpe.
 - J. Quant. Spectrosc. Radiat. Transfer, 72, 139-191(2002).
- 7. Multispectrum analysis of self- and nitrogen-broadening, pressure shifting and line mixing in the v₃ parallel band of ¹²CH₃D.
 - V. Malathy Devi, D. Chris Benner, Mary Ann H. Smith, Curtis P. Rinsland, Linda R. Brown.

- J. Quant. Spectrosc. Radiat. Transfer, 73, 603-640 (2002).
- 8. Self- and N2-broadening, pressure induced shift and line mixing in the v_5 band of $^{12}\text{CH}_3\text{D}$ using a multispectrum fitting technique.
 - V. Malathy Devi, D. Chris Benner, Mary Ann H. Smith, Curtis P. Rinsland, Linda R. Brown
 - J. Quant. Spectrosc. Radiat. Transfer 74, 1-41 (2002).
- 9. Determination of self- and H₂-broadening and shift coefficients in the 2-0 band of ¹²C¹⁶O using a multispectrum fitting procedure.
 - V. Malathy Devi, D. C. Benner, M. A. H. Smith, C. P. Rinsland and A. W. Mantz.
 - J. Quant. Spectrosc. Radiat. Transfer 75, 455-471 (2002).
- 10. Weak ozone isotopic absorption in the 5 mm region from high resolution FTIR solar spectra.
 - A. Goldman, C. P. Rinsland, A. Perrin, J. –M. Flaud, A. Barbe, C. Camy-Peyret, M. T. Coffey, W. G. Mankin, J. W. Hanigan, T. M. Stephen, V. Malathy Devi ad M. A. H. Smith.
 - J. Quant. Spectrosc. Radiat. Transfer 74, 133-138 (2002).
- 11. Nitrogen broadening and shift coefficients in the 4.2 to 5.5-μm bands of CO₂.
 - V. Malathy Devi, D. C. Benner, M. A. H. Smith, and C. P. Rinsland.
 - J. Quant. Spectrosc. Radiat. Transfer 76, 289-307 (2003).
- 12. Absolute intensity measurements of the ${}^{12}C^{16}O_2$ laser bands near $10\mu m$.
 - V. Malathy Devi, D. C. Benner, M. A. H. Smith, L. R. Brown and M. Dulick.
 - J. Quant. Spectrosc. Radiat. Transfer 76, 393-410 (2003).
- 13. Multispectrum analysis of pressure broadening and pressure shift coefficients in the ¹²C¹⁶O₂ and ¹³C¹⁶O₂ laser bands.
 - V. Malathy Devi, D. C. Benner, M. A. H. Smith, L. R. Brown and M. Dulick.
 - J. Quant. Spectrosc. Radiat. Transfer 76, 411-434 (2003).
- 14. A multispectrum analysis of the v_1 band of $H^{12}C^{14}N$: I. Intensities, self-broadening and self-shift coefficients.
 - V. Malathy Devi, D. C. Benner, M. A. H. Smith, C. P. Rinsland, S. W. Sharpe and R.
 - L. Sams.
 - J. Quant. Spectrosc. Radiat. Transfer (to appear in the special issue of JQSRT).

- 15. A multispectrum analysis of the v_1 band of $H^{12}C^{14}N$: II. Air- and N_2 -broadening, shifts and their temperature dependences.
 - V. Malathy Devi, D. C. Benner, M. A. H. Smith, C. P. Rinsland, S. W. Sharpe and R. L. Sams.
 - J. Quant. Spectrosc. Radiat. Transfer (to appear in the special issue of JQSRT).

Oral Presentations:

- Aoaeh, B.; Abebe, Y.; Mantz, A.W.; Ball, C.D.; De Lucia, F.C.; Benner, D.C.; and Smith, M.A.H.: Lead salt semiconductor laser emission lineshape analysis. Presented at the 55th Ohio State University International Symposium on Molecular Spectroscopy, Columbus, Ohio, June 12-16, 2000 (Paper ME06, p. 77).
- Aoaeh, B.; Kolodziejski, N.; Mantz, A.W.; Benner, D.C.; Malathy Devi, V.; Smith, M.A.H.; Ball, C.D.; and De Lucia, F.C.: Measurements of the 1-0 band of carbon monoxide at temperatures between 11 and 296 Kelvins. Presented at the 55th Ohio State University International Symposium on Molecular Spectroscopy, Columbus, Ohio, June 12-16, 2000 (Paper ME07, p. 77)
- Smith, M.A.H.; Rinsland, C.P.; Malathy Devi, V.; Benner, D.C.; Stephen, T.M.; Goldman, A.; and Perrin, A.: High resolution spectra of isotopic ozone in the 5 µm region. Presented at the 55th Ohio State University International Symposium on Molecular Spectroscopy, Columbus, Ohio, June 12-16, 2000 (Paper WG03, p. 167).
- Benner, D.C.; Malathy Devi, V.; Smith, M.A.H.; Rinsland, C.P.; Guelachvili, G.; Picqué, N.; and Brown, L.R.: Temperature dependence of line mixing in the P branch of the v₃ band of methane. Presented at the 55th Ohio State University International Symposium on Molecular Spectroscopy, Columbus, Ohio, June 12-16, 2000 (Paper WG09, p. 170).
- Malathy Devi, V.; Benner, D.C.; Smith, M.A.H.; Rinsland, C.P.; Brown, L.R.; and Sams, R.L.: Nitrogen broadening and shift coefficients in the ν₅ and ν₆ fundamental bands of ¹²CH₃D. Presented at the 55th Ohio State University International Symposium on Molecular Spectroscopy, Columbus, Ohio, June 12-16, 2000 (Paper WG10, p.170).
- Brown, L.R.; Malathy Devi, V.; Benner, D.C.; Smith, M.A.H.; Rinsland, C.P.; and Sams, R.L.: Analysis of self-broadened spectra in the ν₅ and ν₆ fundamental bands of ¹²CH₃D. Presented at the 55th Ohio State University International Symposium on Molecular Spectroscopy, Columbus, Ohio, June 12-16, 2000 (Paper WG11, p. 171).
- Smith, M.A.H.; Rinsland, C.P.; Malathy Devi, V.; Benner, D.C.; and Brown, L.R.: Nitrogen- and self-broadening and shift coefficients in the v₃ fundamental band of ¹²CH₃D. Presented at the 55th Ohio State University International Symposium on Molecular Spectroscopy, Columbus, Ohio, June 12-16, 2000 (Paper WG12, p.171).
- Benner, D.C.; Malathy Devi, V.; Brown, L.R.; Smith, M.A.H.; and Rinsland, C.P.: Line mixing in the triad of ¹²CH₃D. Presented at the 55th Ohio State University

- International Symposium on Molecular Spectroscopy, Columbus, Ohio, June 12-16, 2000 (Paper WG13, p. 172).
- Benner, D.C.; Malathy Devi, V.; and Smith, M.A.H.: Characterization of the v₃ band of methane. Presented at the Sixth Biennial HITRAN Conference, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts, June 19-21, 2000 (Session 3, p. 31).
- Smith, M.A.H.; Malathy Devi, V.; Benner, D.C.; Brown, L.R.; and Sams, R.L.: Air-, N₂- and self-broadening and shift coefficients and line mixing in the v₃, v₅ and v₆ bands of ¹²CH₃D. Presented at the Sixth Biennial HITRAN Conference, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts, June 19-21, 2000 (Paper P21, p. 37).
- Nikitin, A.; Champion, J.P.; Brown, L.R.; Malathy Devi, V.; Benner, D.C.; Smith, M.A.H.; and Tyuterev, V.G.: Line strengths of CH₃D in the region 900 3500 cm⁻¹. Presented at PRAHA2000, the 16th International Conference on High Resolution Molecular Spectroscopy, Prague, Czech Republic, September 3-7, 2000 (Paper D11).
- Mantz, A.W.; Kolodziejski, N.; Aoaeh, B.; Benner, D.C.; Malathy Devi, V.; Smith, M.A.H.; Ball, C.D.; and De Lucia, F.C.: Measurements of the R(0) line in the 1-0 band of carbon monoxide at temperatures between 11 and 296 Kelvins. Presented at PRAHA2000, the 16th International Conference on High Resolution Molecular Spectroscopy, Prague, Czech Republic, September 3-7, 2000 (Paper H26).
- Mantz, A.W.; Kolodziejski, N.; Gobeille, R.; Benner, D.C.; Malathy Devi, V.; and Smith, M.A.H.: Some preliminary results of a simultaneous analysis of an RQ(J,0) Q branch in methyl fluoride at 1475 wavenumbers. Presented at PRAHA2000, the 16th International Conference on High Resolution Molecular Spectroscopy, Prague, Czech Republic, September 3-7, 2000 (Paper H40).
- Smith, M.A.H.: Characterization of line broadening and shift parameters of ozone for spectroscopic databases. Presented at the 56th International Symposium on Molecular Spectroscopy, Ohio State University, June 11-15, 2001 (Paper ME03).
- Mantz, A.W., Smith, M.A.H., Rinsland, C.P., De Lucia, F.C., Benner, D.C., and Malathy Devi, V.: Temperature dependence of air-broadening and shift coefficients in the ¹²C¹⁶O fundamental band. Presented at the 56th International Symposium on Molecular Spectroscopy, Ohio State University, June 11-15, 2001 (Paper ME06).
- Benner, D.C., Malathy Devi, V., Smith, M.A.H., Rinsland, C.P., and Mantz, A.W.: Self-broadening, shifting, and line mixing in the fundamental band of ¹²C¹⁶O. Presented at the 56th International Symposium on Molecular Spectroscopy, Ohio State University, June 11-15, 2001 (Paper ME07).

- Smith, M.A.H., Rinsland, C.P., Mantz, A.W., Benner, D.C., and Malathy Devi, V.: Self-and H₂-Broadening and Shift Coefficients in the 2-0 Band of ¹²C¹⁶O. Presented at the 56th International Symposium on Molecular Spectroscopy, Ohio State University, June 11-15, 2001 (Paper ME09).
- Benner, D.C., Malathy Devi, V., Smith, M.A.H., Rinsland, C.P., Brown, L.R., and Dulick, M.: Broadening and shift coefficients in the ¹²C¹⁶O₂ and ¹³C¹⁶O₂ laser bands. Presented at the 56th International Symposium on Molecular Spectroscopy, Ohio State University, June 11-15, 2001 (Paper ME11).
- Smith, M.A.H.: Infrared spectroscopic parameters of ozone and other atmospheric gases. Presented at the NASA Workshop on Laboratory Spectroscopy Needs for Atmospheric Sensing, San Diego, California, October 23-26, 2001.
- Malathy Devi, V.; Benner, D.C., Smith, M.A.H.; and Rinsland, C.P.: New laboratory measurements of 4.3 and 10 μm CO₂ bands. Presented at the NASA Workshop on Laboratory Spectroscopy Needs for Atmospheric Sensing, San Diego, California, October 23-26, 2001.
- Rinsland, C.P., Malathy Devi, V., Smith, M.A.H., and Benner, D.C.: Infrared laboratory measurements of HCN broadened by N₂ and air. Presented at the Seventh Biennial HITRAN Conference, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts, June 12-14, 2002.
- Malathy Devi, V., Benner, D.C., Smith, M.A.H., Brown, L.R., and Dulick, M.: Spectral line parameters for the ¹²C¹⁶O₂ and ¹³C¹⁶O₂ laser bands. Presented at the Seventh Biennial HITRAN Conference, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts, June 12-14, 2002.
- Smith, M.A.H., and Malathy Devi, V.: Vibrational dependence of broadening and shift parameters for linear molecules. Presented at the Seventh Biennial HITRAN Conference, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts, June 12-14, 2002.
- Benner, D.C., Malathy Devi, V., and Smith, M.A.H.: Effects of line mixing upon spectroscopic line parameters in the R branch of the 1-0 band of CO. Presented at the Seventh Biennial HITRAN Conference, Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts, June 12-14, 2002.
- Smith, M.A.H., Rinsland, C.P., Benner, D.C., and Malathy Devi, V.: Broadening and shift coefficients in the v_3 bands of $^{12}C^{16}O_2$ and $^{13}C^{16}O_2$. Presented at the 57th International Symposium on Molecular Spectroscopy, Ohio State University, June 17-21, 2002 (Paper MH10).
- Smith, M.A.H., Rinsland, C.P., Benner, D.C., Malathy Devi, V., and Dulick, M.: Broadening and shift coefficients in the 3 µm bands of HCN. Presented at the

57th International Symposium on Molecular Spectroscopy, Ohio State University, June 17-21, 2002 (Paper MH05).

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Hampton, Virginia July 10, 2003